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ULTIMATE DISPOSAL OF ADVANCED WASTE TREATMENT RESIDUES

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Ultimate Disposal of Advanced Waste Treatment Residues

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Advanced Waste Treatment processes include methods for removing pollutants such as nutrients, suspended matter, dissolved organics, and salts from waste water and concentrating them in a waste stream. Devising the most economical process for the disposal of the concentrated pollutants is the responsibility of the Ultimate Disposal Research Activities of the Federal Water Pollution Control Administration in Cincinnati. Pollutants that are removed from waste waters must be treated so that they will not interfere with the intended use of the environment. The elements of polluting substances may be placed in the air, either on or under the land, and in the oceans. Preliminary conversion to innocuous or nonextractable forms is necessary. Organic substances may be oxidized to carbon dioxide, water, and nitrogen by dry incineration, wet oxidation at high temperature and pressure, or by soil organisms. Water-soluble salts must be locked up or diluted in the ocean. Other elements should be recovered for chemical values or converted to insoluble precipitates which can be buried or sunk.

Keywords: Waste disposal · Water pollution · Water reclamation · Recovering Phosphates · Calcium oxide · Aluminum hydroxide

ADVANCED waste treatment (AWT) processes have been developed over the past 7 years to remove from waste water those pollutants that are not removed by conventional treatment. Middleton *et al.* have summarized the state of the art of AWT (*1*, *2*). Nearly all of the processes which can be used to purify municipal waste water are also suitable for purifying industrial wastes, and most of them have been tested on pulp and paper mill effluents.

Advanced waste treatment processes being studied include the following:

1. Although not originally included as an AWT process, biological treatment can be substantially improved and under some conditions will remove significant quantities of nutrients.

2. Foam concentration, the first AWT process to be proved on a pilot scale, is practiced in some types of paper mill recovery operation.

3. Coagulation followed by sedimentation is another well known but poorly understood way to remove solids and some soluble materials from waste waters.

4. Facilities for activated carbon adsorption are now being installed for use on a 7.5 MGD (million gallons per day) scale at Lake Tahoe to adsorb soluble organic substances from treated waste water. The ability of activated carbon to remove soluble organic matter from wastes is recognized, but costs are high when treating lignin color in pulp mill effluents.

5. Reverse osmosis, electrodialysis, and ion exchange are rival methods for reducing concentrations of soluble salts. All three are being tested in pilot plants on waste water and have been tried on paper mill effluents.

Whenever waste water is purified to reduce pollution it is probable that the water can be reused. While this concept may be repugnant when applied to sewage many of our rivers that receive municipal wastes are already being reused as water supplies by communities downstream from waste outfalls. Reuse of industrial waters after treatment is, of course, widespread.

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DISPOSAL OF RESIDUES

General

Advanced waste treatment always involves separation and degradation: separation of the good part from the bad part, and degradation of the bad. To this extent it is similar to the processing of the products of farms, mines, or forests. In all of these industries a valuable component, be it wheat, copper, or pulp, must be separated from a component of little or no value, i.e., straw, gangue, or lignin. The low-value residue usually becomes an economic liability and, in the absence of restrictive legislation, may be dumped and become a pollutant. In this country we are deciding that neither water nor air should be used for disposal of noxious substances, but we still approve of controlled burial for the disposal of unwanted organic matter and the use of the atmosphere for the disposal of carbon dioxide. There is also no objection to putting clean water back into the river and, by corollary, there should be no objection to returning salt to the ocean.

In these and other ultimate disposal methods it is important that a pollutant removed from one medium does not cause pollution elsewhere. For example, wet scrubbers very effectively reduce air pollution from furnaces but they produce dirty water which needs further treatment before it is discharged. Likewise, sewage sludge may be disposed of by incineration, but the incineration furnace should be designed to control air pollution.

Before disposal it is ordinarily necessary to render innocuous those substances which have been removed from waste water. This can be done by dilution, as brines from desalting plants are disposed of in the ocean, by locking up in insoluble salts or impermeable caskets, or by chemical conversion to less objectionable substances. In some cases chemical conversion can produce salable products which may help to defray the cost of treatment.

Ocean waters have been used since time immemorial for the disposal of human wastes. Only recently have deleterious effects been noted, and evidence is accumulating that the ocean is neither infinitely large nor rapidly mixed. The principal effect of near shore disposal appears to be a higher fertility of the waters, leading in some cases to better fish environment and in others to objectionable growths of algae and microscopic organisms. Not enough is known about the ecological effects produced by wastes dumped in the sea. There is the possibility that food chains may concentrate biocides, resulting in toxic levels in commercial fish.

Disposal of Solids

Solid materials may be disposed of on the land either as a concentrated fill that is buried, or by dispersing on the surface. In either case, solution of salts or toxic materials in groundwater may produce pollution.

Spreading of liquid sludges on the land surface is one of the oldest and least

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expensive methods of sewage sludge disposal (*3*). Early applications of untreated sludge in an uncontrolled manner produced bad odors and foul seepage to streams, throwing the method into disfavor. However, land spreading is by far the cheapest way for dewatering sludges, and work is being done to improve the operating process. Spreading of large quantities of soluble salts or toxic metals is unsuitable, but most soils have great capacities for dewatering and oxidizing organic matter. The fertilizer value of municipal sludge, in terms of nitrogen, phosphorus, and potassium, is very low compared to synthetic fertilizers, and attempts to market dried sludge for fertilizer have usually failed to show a profit (*4*). The market value of the organic matter in sludges is hard to assess, although its value in improving the quality of sandy soil has been repeatedly demonstrated (*5*).

Fertilizer value aside, land surfaces as low-cost dewatering and oxidizing systems are worth looking at. In many areas farmers are glad to have digested wet sludge spread on their fields, and rivalry to get the wet sludge may eventually give it market value (*6*). Typically, the sludge is spread in a thin layer from a tank truck or sprayed from a large nozzle. The sludge is allowed to dry; puddling is avoided to minimize odors. If plowed frequently, soils can assimilate almost unlimited quantities of organic sludge if it is not poisoned by heavy metals.

Dalton, Stein, and Lynam (*7*) described a potential large-scale application of wet sludge in a Chicago study. They propose a pipeline to transport digested sludge to low-quality land in Cook County and ultimately to land 50 miles south. Morgantown, W. Va., has a federal grant to pipe sludges 5 miles to a strip mine spoil area where it can promote regrowth of plant cover (*8*).

Disposal of Soluble Wastes

Deep-well disposal is attractive for soluble wastes, such as salt solutions, if the underground formation is suitable. The largest scale application of this method is the return of salt brines from petroleum recovery (*9*). Most underground aquifers act as filters and there is great danger that the pores will be clogged if suspended solids are present in the fluid. Water can also be introduced underground by fracturing the rock formations and opening up bedding planes. Here the formations will admit some suspended solids (*10*). Disposal into underground rivers in limestone formations at low pressure is also practiced. Whenever water is placed underground there is danger of polluting nearby groundwater. This is obviously greatest with shallow wells but deep-well injection has also caused pollution in some areas. Changes in surface levels and earthquakes have also been attributed to underground injection of wastes (*11*).

Underground cavities merge with deep wells since cavities may be produced by hydraulic extraction of salt or sulfur as well as by natural solution of limestone. The costs of mined cavities, such as those used for underground storage of fuel gas, are high compared with other means of ultimate disposal. They are more suitable for disposal of radioactive wastes which require continued monitoring. Very large cavities produced by nuclear explosions might be utilized if the geology is favorable (*12*).

CHEMICAL CONSIDERATIONS

A rational analysis of ultimate disposal problems must start with the chemistry of the pollutants. Since polluting substances can be chemically transformed into less obnoxious forms, we should start with the individual elements. Fortunately, we do not have to consider 100 different elements because most are so rare as to constitute pollution problems only by occasional accident. Disposal of radioactive isotopes is not included since the use and disposal of these materials are subject to strict control by the U.S. Atomic Energy Commission. Figure 1 is a periodic chart showing those elements which constitute over 99% of the ultimate disposal problems.

Organic Matter

Organic substances are compounds of carbon with hydrogen, oxygen, and lesser amounts of nitrogen, sulfur, and phosphorus, plus other elements which can be lumped together as ash. The principal problem in disposing of organic matter is, however, not the carbon compounds but the associated water. A 5% primary sludge on a dry-weight basis contains nearly 20 times as much water as all other matter. Waste activated sludge may be concentrated by settling to 2% solids, and at this concentration it contains over 50 times as much water as organic matter. The ultimate form for disposal of organic substances is oxidation to carbon dioxide and water. All organic matter in sewage derived initially from CO_2 in the air and and its return to the atmosphere represents a very small fraction of the daily contributions from the combustion of fuels.

Oxidation by combustion not only disposes of organic matter but also provides heat which can be used. The combustion in an incinerator of 1 lb of dry sludge produces enough heat to evaporate 2–3 lb of water. The actual moisture level that will sustain combustion depends on both the incinerator design and the nature of the sludge. Oily sludges and greases which contain a low percentage of combined oxygen can evaporate 3 lb or more of water. Inerts or ash, of course, contribute no heat of combustion, and a small quantity of heat will be consumed in raising their temperature. Organic matter adsorbed on activated carbon is burned to CO_2 when the carbon is regenerated. Organic matter applied to the soil is oxidized by the soil microorganisms to produce CO_2 and humus. Phosphates and ammonia are adsorbed on soil minerals and utilized by the crops and there may be some denitrification of soluble nitrates.

Wet oxidation by the Zimmerman process is a pressure treatment in the presence of dissolved air (*13*). The high-temperature process operates just below the critical temperature of water, produces a sterile inert sludge, oxidizes the organic substances to aldehydes, acids, and CO_2, and solubilizes nitrogenous compounds. Phosphates can be retained in the solids by adding lime. The low-temperature process hydrolyzes proteins and slimes and produces an easily filterable sludge cake that is sterile but still high in organic matter. The supernatant liquor is rich in amino acids, other soluble nitrogen compounds and phosphates (*14*). In both systems the supernatant liquor is a

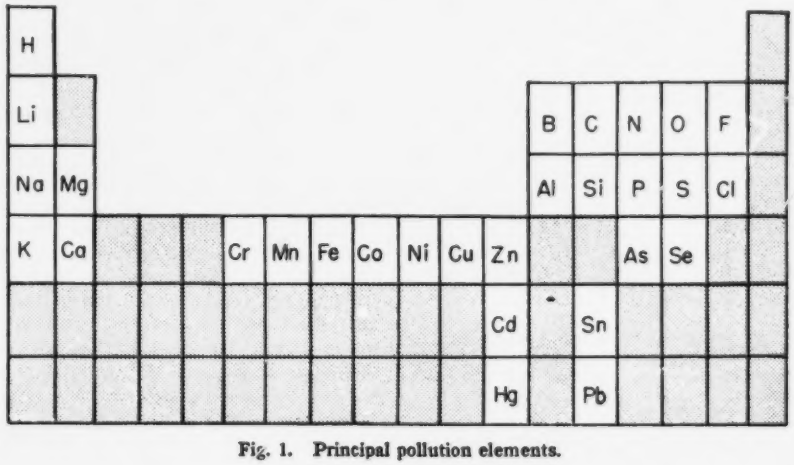


Fig. 1. Principal pollution elements.

foul polluted stream which adds to the load on the biological plant.

Phosphates

Phosphates are the greatest uncontrolled pollutant in waste waters today (*15*). Orthophosphate is a prime nutrient for algal growth in surface waters and present treatment removes only a fraction of the phosphate load in the form of organic sludges. Phosphorus occurs in at least three forms in raw sewage, ortho, condensed, and organic phosphate; normal biological treatment hydrolyzes about 90% of the total phosphate to orthophosphate in secondary effluents (*16, 17*).

Orthophosphates can be precipitated from secondary effluents by treatment with a number of polyvalent cations. Of these, calcium in the form of lime is the least expensive. The precipitate formed is apatite, $Ca_5OH(PO_4)_3$, properly named hydroxyapatite, which is identical to the mineral that forms bones and teeth. This mineral is frequently and erroneously referred to and calculated as tricalcium phosphate, $Ca_3(PO_4)_2$, a substance which does not form in aqueous systems (*18*).

Even at a slightly alkaline pH, waste waters are supersaturated with apatite. However, the crystallization is so slow that weeks or months are required to precipitate substantial quantities of phosphates. By raising the pH, precipitation can be greatly accelerated. The most convenient way to do this is to add hydrated lime, $Ca(OH)_2$. Up to 600 mg/liter of $Ca(OH)_2$ (450 as CaO) are proposed to remove phosphates from secondary effluents at Lake Tahoe (*19*). This quantity of lime raises the pH above 11 and precipitates calcium carbonate and magnesium hydroxide by the same reactions that occur in the cold lime softening of water (Table I). Magnesium hydroxide forms a gelatinous precipitate which aids in clarification and increases the removal of phosphates. In the absence of a clarifying agent, there is danger that some of the colloidal apatite crystals may remain in suspension and redissolve when the effluent is neutralized.

The sludge from waste water lime-treated to remove phosphates contains calcium carbonate, magnesium hydroxide, and organic matter in addition to apatite. Magnesium may be leached out with CO_2 at pH 9.5, the pH of minimum solubility of calcium in the presence of carbonate (*20*). The sludge containing calcium carbonate and apatite can be dried and calcined in a lime kiln to produce calcium oxide, magnesium oxide, and essentially unchanged apatite. The calcium oxide can be slaked to calcium hydroxide and used again to precipitate phosphates. Magnesium oxide ordinarily does not slake or hydrate and remains with the inert fraction.

The extent of lime reuse depends on the hardness of the water. In a hard-water area in southwestern Ohio, lime treatment softens the water, producing more lime than is charged into the process. As a result, the recovered lime must be continuously wasted and might be sold as a low grade of lime. In a soft water area the phosphates remove lime from the process and the apatite can build up in the recycled lime until it reaches a level comparable in strength to a low grade rock phosphate. As such it may have some market value to defray part of the cost of treatment. Figure 2, based on studies in Cincinnati, compares the analysis of recovered lime sludges after repeated reuse in a hard water area and in an area with a moderately soft water (*21*).

At Lake Tahoe the lime will be reused three times before the phosphates and inert substances build up to the point where it must be discarded. At this location there is no market for phosphates, and the waste will be dumped. Fortunately, the phosphate in the ash is insoluble in water and will not leach out into water percolating through the dump. The phosphates, however, do have citrate solubility and could be used for fertilizer if there were a market nearby.

The reuse of lime sludges helps to reduce the cost of phosphate removal but it does not offer a product which can be sold at a profit. Recovered lime may cost nearly as much as commercial lime, and the only savings on chemicals are in the freight. The real savings in lime reuse comes in the reduced disposal problem, since lime sludges accumulate rapidly and are difficult to dispose of.

Aluminum, like other trivalent metals

Table I. Lime Clarification, Recovery, and Reuse

Precipitation

$$3\,Ca^{++} + 4\,OH^- + 3\,HPO_4^{--} \rightarrow Ca_5OH(PO_4)_3 \downarrow + 3\,H_2O$$
$$Ca(OH)_2 + Ca(HCO_3)_2 \rightarrow 2\,CaCO_3 \downarrow + 2\,H_2O$$
$$Mg^{++} + Ca(OH)_2 \rightarrow Mg(OH)_2 \downarrow + Ca^{++}$$

Recarbonation

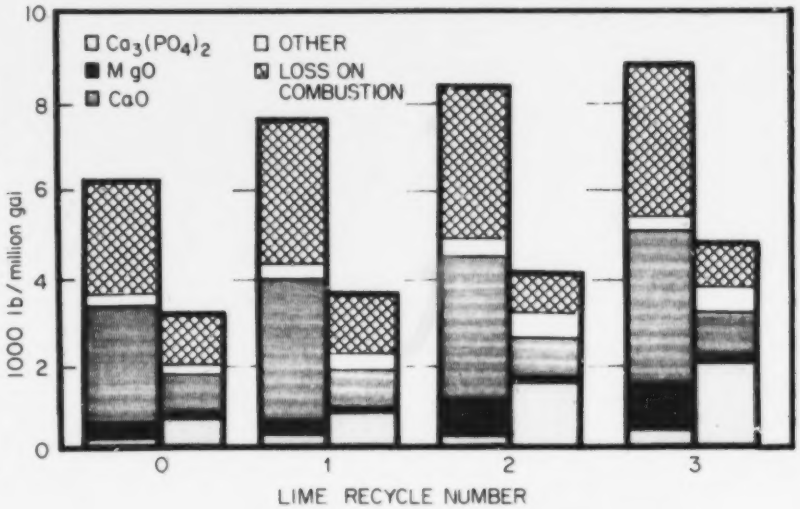
$$Ca^{++} + 2\,OH^- + CO_2 + H_2O \rightarrow CaCO_3 \downarrow + 2\,H_2O$$
$$Mg(OH)_2 + 2\,CO_2 \rightarrow Mg^{++} + 2\,HCO_3^-$$

Recalcination

$$CaCO_3 \xrightarrow{\Delta} CaO + CO_2$$
$$Mg(OH)_2 \xrightarrow{\Delta} MgO + H_2O$$

Hydration

$$CaO \cdot MgO \rightarrow MgO + Ca(OH)_2 + \text{heat}$$



Secondary Effluent Characteristics

	Hard	*Moderately soft*
Ca and Mg hardness, mg/liter $CaCO_3$	410 ± 40	180 ± 30
Alkalinity, mg/liter $CaCO_3$	360 ± 40	85 ± 30
PO_4—P, mg/liter	4 ± 2	14 ± 4
Lime dose, mg/liter CaO M	450	225

Fig. 2. Typical reuse of lime in hard and moderately soft water areas.

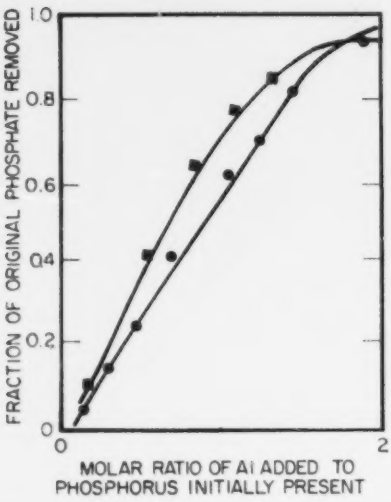


Fig. 3. Removal of phosphate from waste water by aluminum hydroxide.

ions, hydrolyzes in water to form aluminum hydroxide which adsorbs and precipitates phosphates at a nearly neutral pH. Either alum, $Al_2(SO_4)_3 \cdot XH_2O$, or sodium aluminate, $NaAl(OH)_4$, can be used to form the hydroxide by suitable pH adjustment (Table II) (*22*). The exact chemical form of the aluminum phosphate is not known, but for high removal efficiency an Al-to-P ratio of 2:1 is necessary. This corresponds to the formulas $Al(OH)_2AlPO_4$, but a polymeric form is more likely. Figure 3, based on the work of Henriksen (*23*), shows the percent removal of phosphate by aluminum as a function of mole ratio of Al to P.

Aluminum hydroxide will redissolve at high pH to form aluminate ions. Some of the aluminum phosphate is also dissolved but the phosphate ions can be precipitated by calcium as apatite (*24*). Studies are underway at Cincinnati to find the optimum conditions for aluminate recovery. It should be possible to supply a substantial portion of the alkali requirements with lime. Aluminates can be used as a source of aluminum hydroxide for further treatment by neutralizing the alkali. Bicarbonate alkalinity in the waste water may be sufficient, but in some cases additional acid may be required. If some aluminum is lost with the calcium phosphate, both aluminum and acid can be supplied as alum which carries 3 equivalents of acidity per mole of aluminum.

Disposal of aluminum hydroxide sludges from water treatment has been, and will remain, a difficult problem. Sludges rich in organic matter may be difficult to salvage by conversion to aluminate because humic acids will also dissolve. Very small quantities of aluminum hydroxide clog the soil and prevent use of the land for agriculture. Dewatering of aluminum hydroxide is extremely difficult. The only successful method to date has been to freeze the sludge (*25*). This removes the free water and gives an alumina that filters like sand. Freezing is, of course, an expensive process. Incineration of the sludge produces insoluble aluminum phosphates in the ash.

Nitrogenous Compounds

Nitrogenous compounds represent the second greatest uncontrolled pollutant in municipal waste waters (*26*). All these compounds are derived from the atmosphere and eventually return to it in the nitrogen cycle. All the biologically significant forms of nitrogen are interconvertible; they should be reported on a common basis such as milligrams of ammonia nitrogen, or nitrate nitrogen, etc., per liter. Nitrogenous compounds, except N_2 in the atmosphere, stimulate the growth of algae and lead to nuisance conditions. Blue-green algae also have the ability to "fix" nitrogen from the air, producing organic nitrogen which then enters the nitrogen cycle. Nitrogenous compounds in general are good fertilizers when applied to the land, where nitrogen deficiencies frequently limit crop production, but they are bad fertilizers in surface waters if they contribute to excessive growth of algae and weeds.

Organic compounds of nitrogen can be burned to nitrogen gas. Furnaces should be carefully designed to minimize production of oxides of nitrogen, but in most cases fixation of atmospheric nitrogen will be much greater than nitrogen oxides from organic compounds. Organic nitrogen compounds are hydrolyzed to ammonia by microorganisms under aerobic or anaerobic conditions.

Ammonia is the most common form of nitrogen in waste waters, averaging about 15 mg/liter as N (*17*). If these waters are to be reused, the ammonia level must be brought down below 1 mg/liter. Chlorination to the breakpoint converts ammonia to nitrogen gas, but 10 mg of chlorine is required per milligram of ammonia nitrogen (*27*). Breakpoint chlorination of the typical secondary effluent would cost 5–10¢ per 1000 gal.

At the pH values of normal secondary effluents, ammonia is present almost entirely as ammonium ions. Above pH 10 most of the ammonia is present as dissolved gas. Aeration can remove gaseous ammonia from water but the equilibrium is very strongly in favor of the water phase. About 300 ft^3 of air are required per gallon of water at room temperature and an initial pH 10.6. At Lake Tahoe the high pH will be obtained by lime treatment to remove phosphates, and air stripping will be carried out in large cooling towers. The cost of air stripping is estimated to be under 2¢ per 1000 gal (*19*).

Salts

The only acceptable sites for disposal of salts in solution are the ocean or salt lakes. In some areas salt brines are collected in lagoons and discharged to the river during a spring flood. With proper control, limited quantities of salts might not pollute some rivers at high flow.

SUMMARY

The intelligent disposal of wastes can only be accomplished by a systematic study of the chemistry of the substances involved. Under appropriate conditions, wastes can be disposed of economically on or under the land, into the oceans or into the air, but in every case precautions must be taken to control pollution.

The cost of disposing of sludges and brines is largely associated with the water content. The high costs of dewatering and drying make pipeline transport of wet sludges into the ocean or onto the land economically attractive.

Organic wastes will be oxidized, either by microorganisms in the water or on the land, or by heat in an incinerator to CO_2. Phosphates must be removed as calcium, aluminum, or iron precipitates. Calcium may be recovered as reburned lime, and aluminum as aluminates. In these recovery processes, the phosphate eventually comes out as apatite, the natural calcium phosphate mineral, which has low solubility in ground water but is a potential raw material for fertilizer production.

Nitrogenous compounds are converted to nitrogen gas by incineration or biological denitrification. Ammonia can be blown off at high pH or can be adsorbed on soil minerals. Soluble salts must go to the sea, to salt lakes, or salty underground aquifers.

Table II. Phosphate Removal with Al^{+3}

Influence of pH on Al^{+3} Form

$$Al^{+++} \underset{}{\overset{+3OH^-}{\rightleftharpoons}} Al(OH)_3\downarrow \overset{+OH^-}{\rightleftharpoons} Al(OH)_4^-$$

pH 3 pH 7 pH 12

Phosphate Removal

$$2\,Al(OH)_3 + HPO_4^- \rightarrow AlPO_4 \cdot Al(OH)_3\downarrow + 2\,OH^- + H_2O$$

Solubilization

$$3\,AlPO_4 \cdot Al(OH)_3\downarrow + 5\,Ca(OH)_2 + 6\,NaOH \rightarrow 6\,NaAl(OH)_4 + Ca_5OH(PO_4)_3\downarrow$$

Every waste water purification scheme must look beyond the initial purification process, be it activated sludge, alum clarification, or reverse osmosis, to include in the total costs the ultimate disposal of the residual pollutants.

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